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Comparison of spatial patterns of pollutant distribution with CMAQ predictions

Sharon B. Phillips[a,*], Peter L. Finkelstein[b]

[a]United States Environmental Protection Agency, Office of Air and Radiation, Office of Air Quality Planning and Standards, Mail Drop D243-01, 109 T.W. Alexander Drive, RTP, NC 27711, USA
[b]National Oceanic and Atmospheric Administration, Atmospheric Sciences Modeling Division, in partnership with the United States Environmental Protection Agency, RTP, NC 27711, USA

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Abstract

To evaluate the Models-3/Community Multiscale Air Quality (CMAQ) modeling system in reproducing the spatial patterns of aerosol concentrations over the country on timescales of months and years, the spatial patterns of model output are compared with those derived from observational data. Simple spatial interpolation procedures were applied to data from the Clean Air Status and Trends Network (CASTNet) and Speciation Trends Network (STN) monitoring networks. Species included sulfate PM, total nitrate ($NO_3^- + HNO_3$), and ammonium PM. Comparisons were made for the annual average concentrations for 2001, and for one lunar month (4 weeks), where the month chosen for each species represents the highest concentrations of the year. Comparisons between the modeled and interpolated spatial patterns show very good agreement in the location and magnitude of the maxima and minima, as well as the gradients between them. Some persistent biases are identified and noted. Limitations on our ability to describe the spatial pattern from sparse data as well as the limitations of the networks are briefly discussed.
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1. Introduction

Air pollution models are used as a deterministic tool in order to reduce the degree of ambiguity in $PM_{2.5}$ ambient concentrations regionally, nationally, and globally and to predict future air quality (Seigneur, 2001; Boylan et al., 2002). One indication of model performance is the comparison of spatial patterns of pollutants, either as concentration or deposition, predicted by an air quality model with spatial patterns derived from measurements. If the spatial patterns produced by the air quality model are similar to the patterns produced from the observations in shape, location, and magnitude, this can add to our confidence in the model's results. Utilizing these spatial patterns, one can also assess the model's ability to simulate the complex atmospheric interactions/exchanges between various pollutants at the regional scale. However, deriving the spatial patterns from measured pollutant data is not always trivial. Monitoring networks are spatially and temporally sparse, and frequently biased toward certain types of land-use. There may also

*Corresponding author. Fax: +1 919 541 0044.
E-mail address: phillips.sharon@epa.gov (S.B. Phillips).

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be measurement biases among monitoring networks. These biases are considered during the selection of monitoring networks assumed to be representative of the modeled $PM_{2.5}$ species data. It should be noted that spatial modeling and analysis should be viewed as an additional model evaluation tool among the many statistical techniques assessment that are available (Eder and Yu, 2006); however, these techniques alone are not sufficient by themselves to completely evaluate the performance of a complex modeling system. It is well known that model predictions can never perfectly match observations. Results should be interpreted with special caution in areas with extreme topography or in urban settings.

In this paper, we compare the observed spatial patterns with those predicted by the United States Environmental Protection Agency's Models 3/Community Multiscale Air Quality (CMAQ) modeling system for the year 2001. We will consider the grid-averaged concentrations of sulfate (SO_4^{2-}), total nitrate ($NO_3^- + HNO_3$), and ammonium aerosols (NH_4^+) in this analysis.

2. Model configuration

An annual 2001 simulation consisting of a Lambert conformal horizontal domain at 36 km × 36 km model grid cells with 14 vertical layers over the continental United States was conducted utilizing the Models-3/CMAQ modeling system (version 4.4) (Byun and Ching, 1999). Initial and boundary conditions were provided by a global 3D chemistry/transport model, the Goddard Earth Observing System (GEOS-CHEM) from the NASA Data Assimilation Office, at a resolution of 2° latitude by 2.5° longitude. Year-specific 2001 meteorological data provided by the Fifth-Generation NCAR/Penn State Mesoscale Model (MM5) version 3.6.1 (Grell et al., 1994; McNally, 2003) were used and processed by the Meteorology–Chemistry Interface Processor (MCIP) version 2.3 (Otte, 2004; Byun et al., 1999) for model-ready inputs. Anthropogenic emissions were provided by the 1999 National Emissions Inventory (NEI) (http://www.epa.gov/ttn/chief/net/index.html) and projected to 2001, which includes monitoring data from hour-specific emissions for electric generating units. Biogenic emissions were obtained from the Biogenic Emissions Inventory System (BEIS) model version 3.12 (http://www.epa.gov/asmdnerl/biogen.html). A complete description of CMAQ and its characteristics/attributes can be found in the *Introduction of this Special Atmospheric Environment Issue*. A description of the aerosol component can be found in Binkowski and Roselle (2003) and Byun and Schere (2006).

3. Observations and model predictions

Spatial plots of annual average observed and predicted SO_4^{2-}, NH_4^+, and NO_3^- for 2001 are considered in this study. After exploring the development of reliable spatial models for the monitoring data using a variety of gridding approaches (e.g. kriging, nearest neighbor, natural neighbor, polynomial regression, etc.), we chose the radial basis functions interpolation procedure for the SO_4^{2-}, NH_4^+, and NO_3^- fields (Hardy, 1990; Carlson and Foley, 1991). Kriging interpolation was not used because a good variogram model could not be defined over the entire study domain. However, analogous to the kriging variograms are the radial basis kernel functions ($B(h)$) utilized in this paper. When interpolating to a grid cell, the kernel functions define the set of weights to apply to the data points (Surfer® 8, 2002). A multiquadric method was used to fit the species data to produce a smooth surface:

$$B(h) = \frac{1}{\sqrt{h^2 + R^2}} \quad \text{(inverse multiquadric)}, \tag{1}$$

where h is the anisotropically rescaled relative distance from the point to the node, and R^2 is the smoothing factor specified as $R = 0.2/25 \times n$, where n is the number of data points.

Data from CASTNet and STN monitoring networks were combined because their measurement techniques for SO_4^{2-} and NH_4^+ are reasonably similar, although there are some minor differences. Because no two networks (e.g. STN, IMPROVE, CASTNet) measure nitrogen aerosols (because of problems in the partitioning of nitric acid and nitrate PM) in the same way, we chose to use total nitrate ($NO_3^- + HNO_3$) reported by CASTNet for the spatial analysis of nitrogen.

STN data are collected once every 3 or 6 days, depending on the site. CASTNet data are collected as weekly integrated samples. To overcome this difference, 28 day averages (lunar months) were constructed from all networks, with the start date corresponding to the CASTNet sampling schedule. Sites were used if more than 75% of the data for the month were available.

Likewise, model outputs of sulfate, total nitrate, and ammonium were averaged to get monthly and annual totals, and then interpolated to the same grid used for the observations to stay in the latitude/longitude system, utilizing the nearest-neighbor interpolation method. This method is used since the model outputs are on equally spaced grids. The method assigns the value of the model output value to the nearest observation grid cell (Surfer® 8, 2002).

4. Results

4.1. Sulfate

Examination of 2001 annual observed and predicted sulfate spatial patterns reflect favorably on CMAQ's predictive skill (Fig. 1(a) and (b)). The region of maximum concentrations predicted by the model is similar to the interpolated pattern from the data both in location and in magnitude. The east–west gradient is similar within the limited resolution of the data. The only difference seems to be lower modeled concentrations in southern California and the Pacific Coast. In Fig. 1(c), a spatial plot of absolute bias is shown for annual 2001 sulfate, with positive bias (predicted−observed) in red and negative bias in blue. CMAQ simulated annual SO_4^{2-} remarkably well with slight over- and under-predictions ($\pm 0.5\,\mu g\,m^{-3}$) in the east, and under-predictions (-0.5 to $-1\,\mu g\,m^{-3}$) in the Central and Western US (excepting California). Bearing in mind that observational data in the Central and Western US is sparse, limited credibility should be attached to the comparisons in those areas.

In Figs. 2(a) and (b), we show a selected summer case (lunar month average, 17 July 2001–13 August 2001) in order to evaluate model performance given that concentrations of sulfate PM are greater in the summer. Comparison of spatially derived SO_4^{2-} predictions versus observations shows that the model closely replicates the observed patterns of SO_4^{2-}. CMAQ performed quite well in capturing the location of maximum particulate sulfate in Indiana, Ohio and into the borders of Kentucky, West Virginia and Pennsylvania. The model predicted the magnitude and gradients of SO_4^{2-} well in the east, especially the gradients in Georgia, Alabama, Louisiana, and Arkansas. Isopleth contours in the Central US are questionable due to the absence of SO_4^{2-} measurements in this region, although these predicted gradients and variations of SO_4^{2-} PM closely match observations in Missouri, Arkansas, Kansas, and Oklahoma. However, there is an observed peak of SO_4^{2-} in southern California (Los Angeles), which the model does not predict. There are also differences ($0.5–1.0\,\mu g\,m^{-3}$) between observed and model values along the Pacific coast.

4.2. Total nitrate

Annual averages of total nitrate ($NO_3^- + HNO_3$) observed at CASTNet sites and predicted by CMAQ are given in Fig. 3(a) and (b). The patterns are very similar, with observed and predicted maxima of the same magnitude in Indiana and Ohio, and secondary maxima in southeastern Pennsylvania and southern California. The east–west gradients are also quite similar. The total nitrate bias plots show an annual over-prediction (upto $3\,\mu g\,m^{-3}$) in most of the Eastern US (Fig. 3c). Typically in the west, CMAQ under-predicts total nitrate ($\sim -2\,\mu g\,m^{-3}$), whereas in central and southern California nitrate is over-predicted.

Fig. 4 shows a lunar month average of predicted and observed total nitrate for the period 2–29 January 2001. This sample period was chosen because concentrations of total nitrate PM are higher in the winter. Comparison of spatial patterns reveals that the model performed reasonably well, particularly in the east. The model captured the peaks observed in Illinois, Indiana, and Ohio, although displaced in location and magnitude to the Southwest. The model also correctly simulated the location of the observed total nitrate peak in Pennsylvania, however predicting approximately $1\,\mu g\,m^{-3}$ less than observations. The model over-predicts total nitrate in the Southeast (e.g. Georgia, North and South Carolina, Florida, Alabama, and Mississippi) and in the Central US (Louisiana, Arkansas, Oklahoma, and Texas). The model shows the ability to correctly predict the magnitude of total nitrate in southern and central California. Although the observed CASTNet data do not show the increased concentrations in central California (as a result of the placement of CASTNet monitoring sites at high elevation sites), the predictions are believed to be correct, given that very high concentrations for aerosol nitrate have been found in the urban/valley regions by the STN network. The over-prediction of total nitrate in the winter indicates that nitric acid concentrations may also be over-predicted. Nitric acid production and resulting destruction pathways continue to be examined by

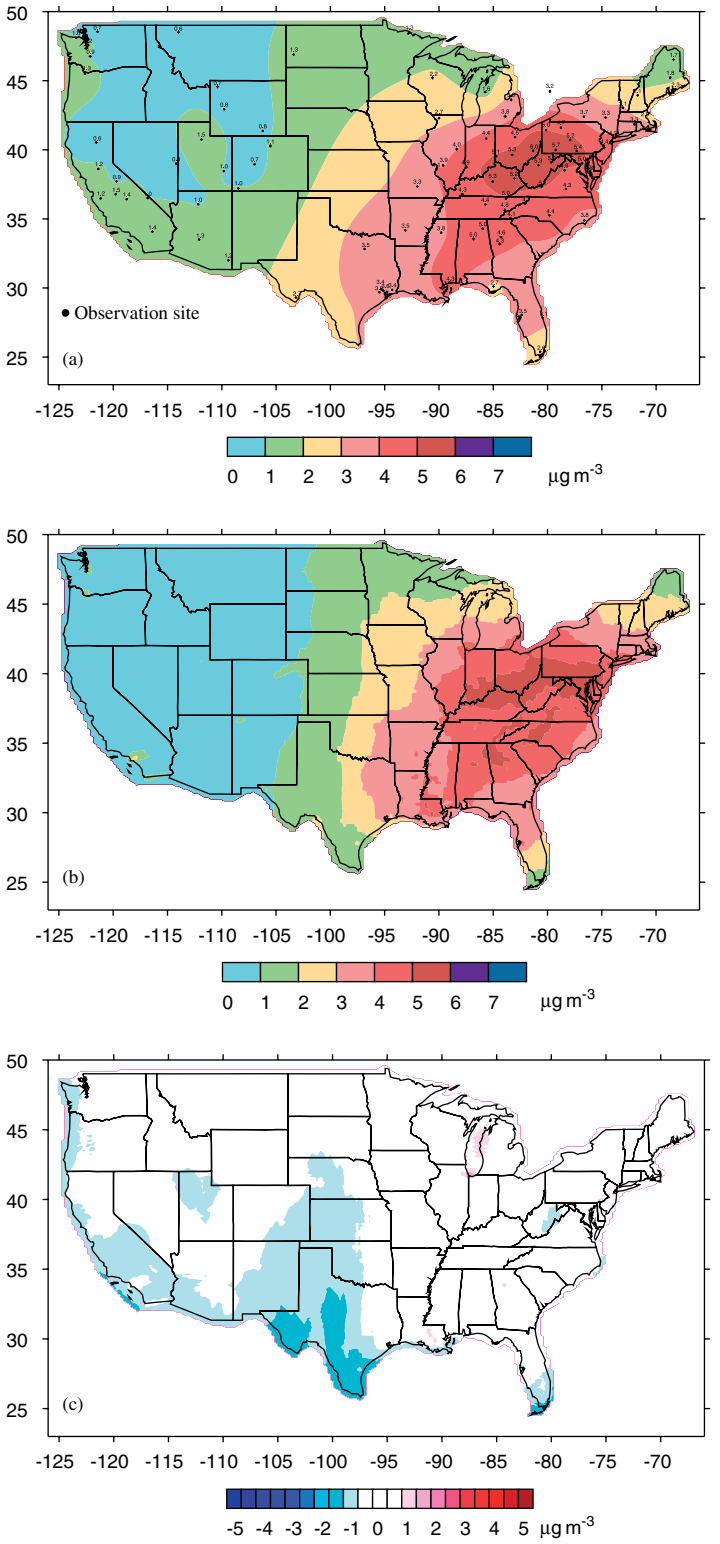

Fig. 1. (a) Spatial plot of observed 2001 annual sulfate from STN and CASTNet; (b) spatial plot of predicted 2001 annual sulfate from CMAQ; (c) bias spatial plot of observed 2001 annual sulfate from STN and CASTNet minus predicted 2001 annual sulfate from CMAQ.

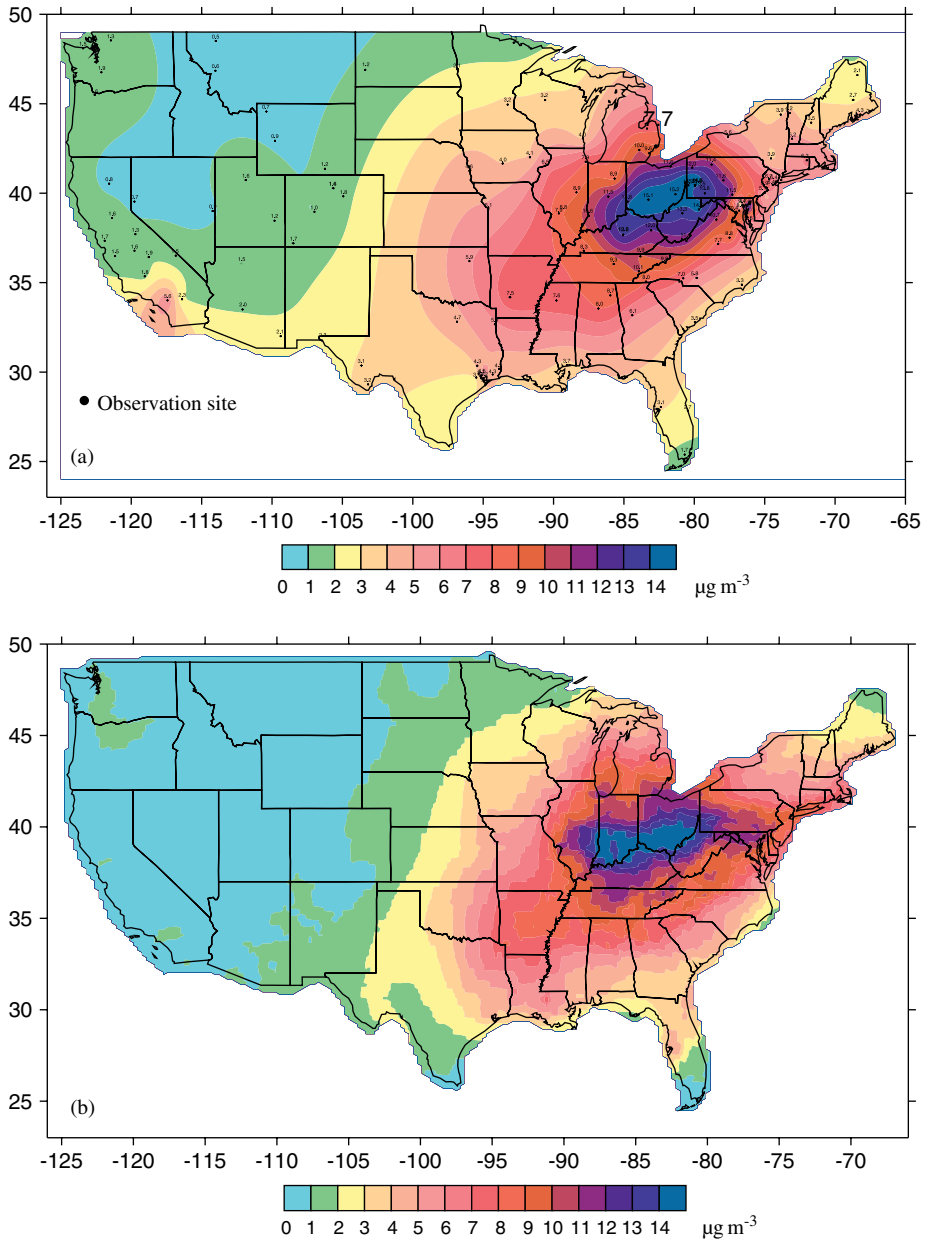

Fig. 2. Lunar month average, 17 July 2001–13 August 2001: (a) spatial plot of observed sulfate from STN and CASTNet; (b) spatial plot of predicted sulfate from CMAQ.

the scientific community, i.e. improvements are being made to the daytime versus nighttime nitric acid formation reactions. The rate of nitric acid dry deposition is also being studied as a possible cause of over-prediction. Excessive ammonia emissions within the model could also contribute to an overestimation of nitrate PM from an imbalance in the conversion of excessive nitric acid to nitrate PM.

4.3. Ammonium

Predicted and observed annual patterns of NH_4^+ are presented in Figs. 5(a) and (b). Observed maxima in the upper midwest, in southern Pennsylvania, and in northern Georgia are captured by CMAQ. They differ in that CMAQ also predicts a secondary maximum in eastern North Carolina that is not observed by the stations in the area. Spatial

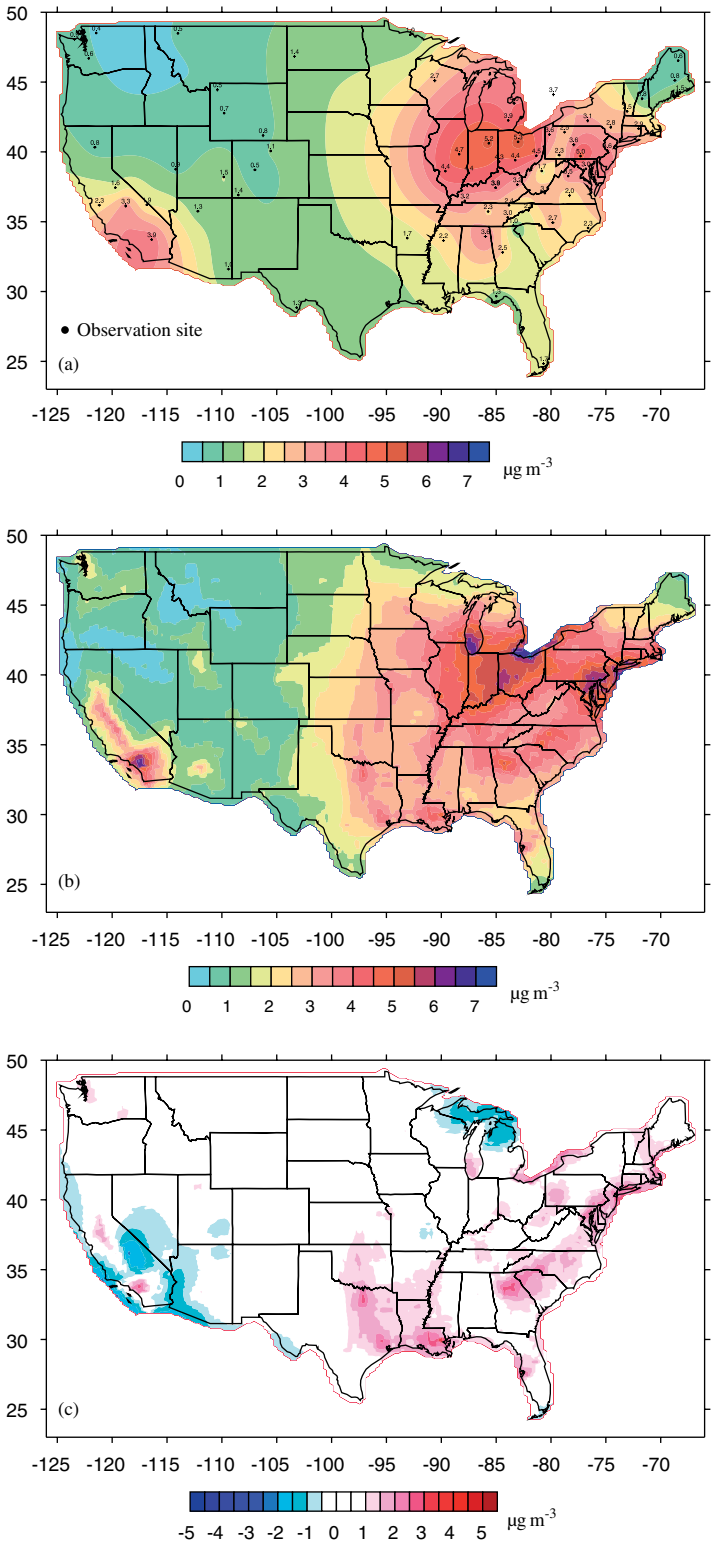

Fig. 3. (a) Spatial plot of observed 2001 annual total nitrate ($NO_3^- + HNO_3$) from CASTNet; (b) spatial plot of predicted 2001 annual total nitrate ($NO_3^- + HNO_3$) from CMAQ; (c) bias spatial plot of observed 2001 annual total nitrate ($NO_3^- + HNO_3$) from CASTNet minus predicted 2001 annual total nitrate ($NO_3^- + HNO_3$) from CMAQ.

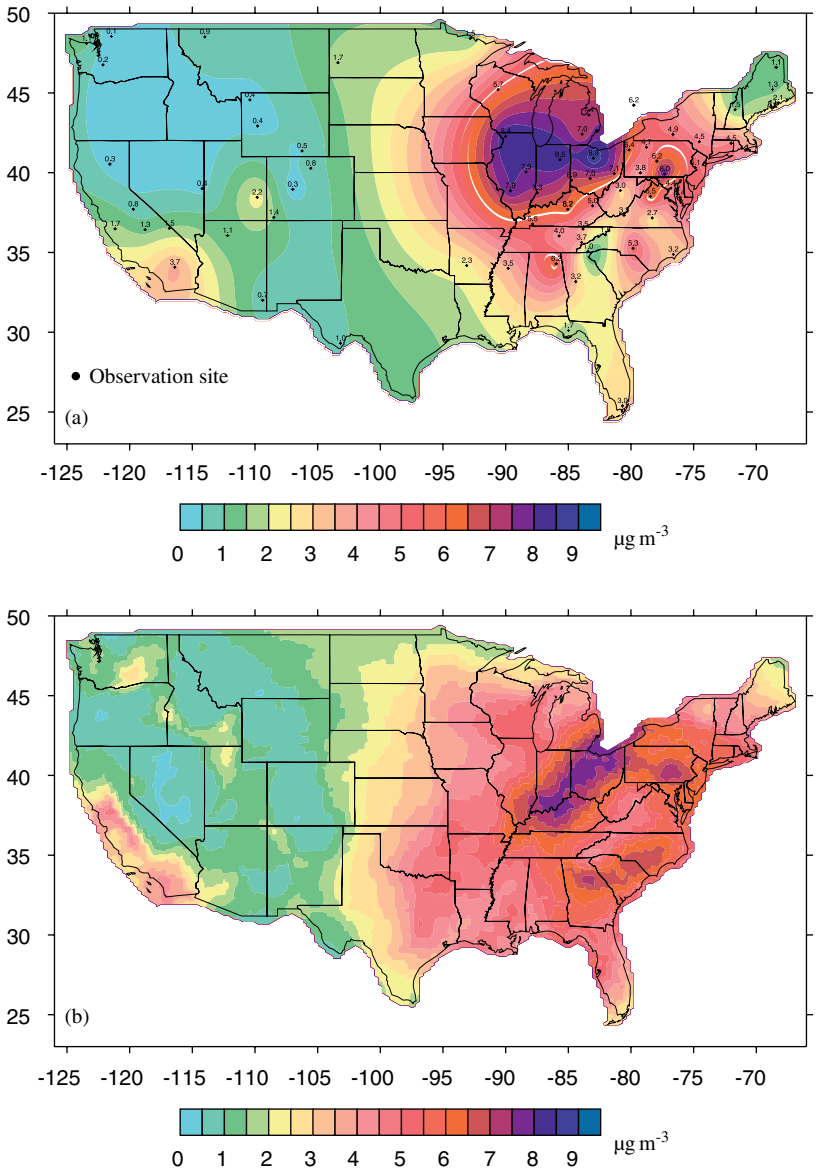

Fig. 4. Lunar month average, 2–29 January 2001: (a) spatial plot of observed total nitrate ($NO_3^- + HNO_3$) from CASTNet; (b) spatial plot of predicted total nitrate ($NO_3^- + HNO_3$) from CMAQ.

patterns in the west are quite similar, although one observing station measured high levels in Salt Lake City that are not predicted by the model. These differences are more clearly seen in Fig. 5(c). In the east, the spatial bias plot shows that CMAQ slightly over-predicts ammonium by approximately 0.5–1 $\mu g\,m^{-3}$. The model performed extremely well in the west, revealing slight differences in observations and predictions in only two small areas in California.

Fig. 6 shows a spatial comparison of observed and predicted NH_4^+ for a summer lunar month average, 17 July 2001–13 August 2001. This lunar month average, like sulfate, is the period of peak ammonium PM. The model does quite well in capturing the spatial gradients and peaks in the east. CMAQ correctly places maxima in Indiana, Ohio and Pennsylvania, but predicts smaller concentrations across most of Indiana and Ohio than were observed. Predicted NH_4^+ concentrations in West Virginia are shown to be low compared to predictions in bordering states and observations, indicating a possible emission input problem in that

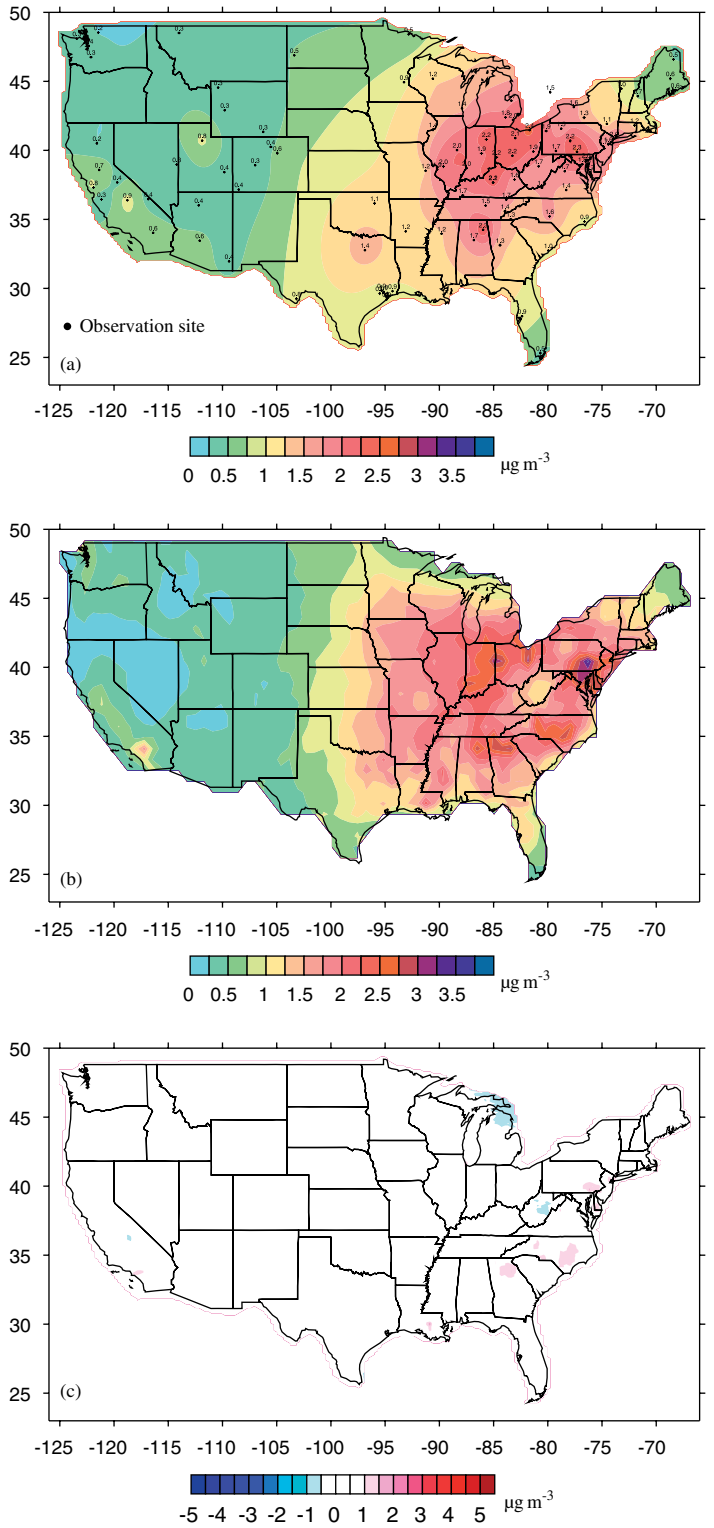

Fig. 5. (a) Spatial plot of observed 2001 annual ammonium from STN and CASTNet; (b) spatial plot of predicted 2001 annual ammonium from CMAQ; (c) bias spatial plot of observed 2001 annual ammonium from STN and CASTNet minus predicted 2001 annual ammonium from CMAQ.

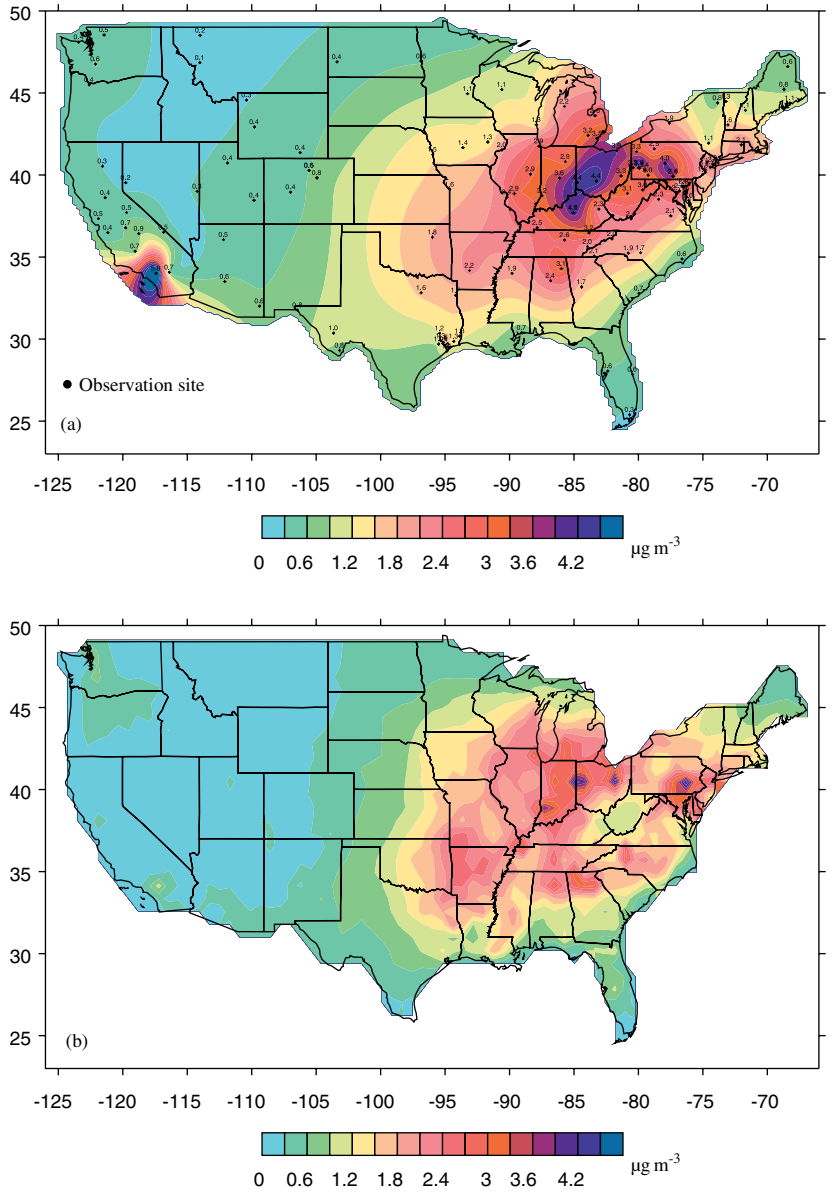

Fig. 6. Lunar month average, 17 July 2001–13 August 2001: (a) spatial plot of observed ammonium from STN and CASTNet; (b) spatial plot of predicted ammonium from CMAQ.

state. In southern California, an observed peak of NH_4^+ ($\sim 5.6\,\mu g\,m^{-3}$) is shown, while the model predicted concentrations in the range of only $0.6–1.5\,\mu g\,m^{-3}$, a significant problem probably due to missing sources in the emissions inventory.

5. Summary

The spatial patterns of sulfate, total nitrate, and ammonium predicted by CMAQ are very similar to the spatial patterns extracted from the observations, in both location and magnitude, especially in the east. Noting that in the Central and Western US, observational data are limited compared to the east, hence, comparisons in these areas should be viewed with caution.

The model predicted the magnitude and gradients of SO_4^{2-} well in the east for both the annual and summer cases presented in this paper. CMAQ simulated annual SO_4^{2-} PM extremely well in the

Eastern and Western US. Spatially derived SO_4^{2-} predictions versus observations show that the model captured the observed patterns and peaks of SO_4^{2-}. CMAQ performed less well for total nitrate when compared to CASTNet observations. Overall, the model tended to over-predict nitrate concentrations over the continental US, although the maxima and spatial gradients are well simulated. These differences are believed to be a result of excessive ammonia where the partitioning between nitrate PM and nitric acid is sensitive to ammonia availability (Stockwell et al., 2000). Likewise, CMAQ performed well for ammonium PM when compared to annual and lunar month 8 (17 July 2001–13 August 2001) spatial maps developed from CASTNet and STN observations. These similarities suggest that CMAQ appears to be doing a credible job of predicting these air pollutant concentrations of these three air pollutants on a regional scale.

This paper considered only spatial analyses of annual lunar month averages, when concentrations were greatest, for three particular $PM_{2.5}$ species. However, spatial analyses for each lunar month of the entire 2001 year have been performed, although not presented here due to page limitations. Overall, the results presented in this paper reflect the findings from all the lunar month averages for each pollutant.

Future research should consider looking into a more robust statistical analysis of spatial interpolation measurements along with bias adjustment. This analysis should include quantifying uncertainties introduced by spatial interpolation techniques of observed measurements as well as model predictions. The use of other monitoring networks should be explored. Case studies involving transport of pollutants should also be undertaken. Techniques are needed to quantify the degree of agreement between predicted and observed maps and to judge the performance of different models against a common data set.

This analysis used numerical interpolation procedures to produce the maps of observed pollutant concentrations. This assumes that the pollutant field is continuous, slowly varying, and without discontinuities between monitoring stations. The validity of these assumptions, in the light of mountainous terrain or large local pollutant sources can be questioned; the maps should be used with caution in these areas, as well as in areas where there are very large distances between monitoring stations. Using strictly numerical interpolation techniques also deprives us of any numerical measure of uncertainty in the maps. Existing statistical techniques, such as kriging, offer some estimate of interpolation uncertainty, but they have their own set of assumptions that are not always met in the real world. Recent developments in spatial statistics offer hope that some of these problems can be overcome, and that robust spatial interpolation procedures with appropriate error estimates applicable to the air pollution field will be available soon.

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References

Binkowski, F.S., Roselle, S.J., 2003. Models-3 Community Multiscale Air Quality (CMAQ) model aerosol component 1. Model description. Journal of Geophysical Research 108 (D6), 4183.

Boylan, J.W., Odman, M.T., Wilkinson, J.G., Russell, A.G., Doty, K.G., Norris, W.B., McNider, R.T., 2002. Development of a comprehensive, multiscale "one-atmosphere" modeling system: application to the Southern Appalachian Mountains. Atmospheric Environment 36, 3721–3734.

Byun, D.W., Ching, J.K.S., 1999. Science Algorithms of the EPA Models-3 Community Multiscale Air Quality (CMAQ) Modeling System, EPA Report 600/R-99/030, Washington DC, Office of Research and Development, US Environmental Protection Agency. http://www.cmascenter.org/ (available from US EPA, ORD,Washington, DC 20460).

Byun, D.W., Schere, K.L., 2006. Review of the governing equations, computational algorithms, and other components of the Models-3 Community Multiscale Air Quality (CMAQ) modeling system. Applied Mechanics Reviews, in review.

Byun, D.W., Pleim, J.E., Tang, R.T., Bourgeois, A., 1999. In: Byun, D.W., Ching, J.K.S. (Eds.), Meteorology–Chemistry Interface Processor (MCIP) for Models-3 Community Multiscale Air Quality (CMAQ) modeling system–Ch12 in Science Algorithms of the EPA Models-3 Community Multiscale Air Quality (CMAQ) Modeling System, EPA-600/R-99/030. Office of Research and Development, US Environmental Protection Agency, Washington, DC (available from US EPA, ORD, Washington, DC 20460).

Carlson, R.E., Foley, T.A., 1991. Radial basis interpolation methods on track data, Lawrence Livermore National Laboratory, UCRL-JC-1074238.

Eder, B.K., Yu, S., 2006. A performance evaluation of the 2004 release of Models-3 CMAQ. Atmospheric Environment, in press, doi:10.1016/j.atmosenv.2005.08.045.

Grell, G., Dudhia, J., Stauffer, D., 1994. A Description of the Fifth-Generation Penn State/NCAR Mesoscale Model (MM5), NCAR/TN-398 + STR. National Center for Atmospheric Research, Boulder, CO, 138pp (updates can be found at http://www.mmm.ucar.edu/mm5/mm5v3/v3model.html).

Hardy, R.L., 1990. Theory and applications of multiquadric–biharmonic method. Computers & Mathematics With Applications 19, 163–208.

McNally, D., 2003. Annual application of MM5 for calendar year 2001. Topical Report, US Environmental Protection Agency, March 2003.

Otte, T.L., 2004, What's new in MCIP2? Preprints. In: Third Annual Models-3 User's Conference, 18–20 October, Chapel Hill, NC, Community Modeling and Analysis System, CD-ROM 7.2. (available from CMAS, c/o Carolina Environmental Program, Bank of America Plaza, CB #6116, 137 E. Franklin St., Chapel Hill, NC 27599).

Seigneur, C., 2001. Current status of air quality models for particulate matter. Journal of the Air and Waste Management Association 51, 1508–1521.

Stockwell, W.R., Watson, J.G., Robinson, N.F., Steiner, W., Sylte, W.W., 2000. The ammonium nitrate particle equivalent of NO_x emissions for wintertime conditions in Central California's San Joaquin Valley. Atmospheric Environment 33, 4477–4493.

Surfer® 8. 2002. User's Guide: Contouring and 3D Surface Mapping for Scientists and Engineers. Golden Software, Inc., 809 14th Street, Golden, Colorado 80401-1866, USA.